

TRIP REPORT - DISCUSSIONS ON THE ASPECTS OF FILTRATION
AND DECONTAMINATION OF RADIOACTIVE EXHAUST AIR

November 12, 1958

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Reactor Modification Design Operation
FACILITIES ENGINEERING OPERATION

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The specific purpose of the subject trip was to obtain as much information as available from the national laboratories on methods and quantitative results of programs which have been developed relative to the decontamination of large volumes of process and/or potentially contaminated ventilation air. In addition, visits were made to selected vendors for the purpose of obtaining up to date information on some of the more critical components of the system proposed under Project CG-791.

The information obtained is presented in the chronological order of the trip.

Argonne National Laboratory, Lemont, Illinois - October 7, 1958

ANL was visited to observe and discuss the operation of a horizontal caustic scrubber in removing radioactive halogens from an air stream prior to release from the exhaust stack. Experimental testing of the horizontal scrubber had previously been reported in ANL-5015 and 5429.(1,2)

W. J. Mecham, ANL
R. C. Liimatainen, ANL
R. W. Kessie, ANL

E. L. Etheridge, HAPO
R. C. Walker, HAPO
M. R. Schneller, AEC, HOO

The horizontal scrubber is in line in an off gas and cell ventilation exhaust system of a pilot plant fuel element dissolver process. The dissolver process consists of adding interhalogen compounds, bromine trifluoride and bromine pentafluoride directly to the uranium slugs. Consequently, there is a high concentration of halogens in the off gas. The exhaust air system is shown in Figure 1.

The scrubber is a welded steel plate box, four feet square and seventeen feet long, with connections at each end for ductwork. It has three successive spray stages, each spray stage consisting of a bank of four hollow spray nozzles impinging on cast iron venturi throat pieces mounted in the stage dividers.

Downstream from the last spray stage is a baffle section followed by a four-inch bed of 250-micron monel fiber mesh to provide de-entrainment. A 50 gpm, recirculating, 10 percent potassium hydroxide solution is used as the spray liquid for a 6000 cfm air flow.

The exhaust gas system operates very well and has been in operation for approximately three years. They estimate that approximately 90 percent of the halogens are removed in the eight-inch vertical scrubber and an additional 90-95 percent halogen removal in the horizontal scrubber section making an overall halogen removal efficiency of 99 1/2 percent. No additional efficiency determinations have been made since the equipment has been put into operation.

Observation of the horizontal scrubber in operation shows that a very fine spray is produced by impingement on the venturi throat pieces. The fine spray is carried along by the air flow to next stage dividers where it is removed from the air stream by impinging on the dividers. Of particular interest was the de-entrainment section. Looking into the scrubber through a viewing window just after the de-entrainment section showed no droplets were visible and no indications that droplets had been formed.

ANL is quite satisfied with the performance and operation of the horizontal scrubber. They are presently fabricating a second scrubber. The two scrubbers are identical with the exception that the second shell is 304 stainless steel. Of interest was a corrosion test performed in a 10 percent KOH and 0.3 percent Na_2NO_2 caustic spray for a period of 1.8 years. The results are listed below:

Metal	Corrosion Loss/Yr.
304 SS	3×10^{-4} mils
Monel	0.03 - 0.18 mils
Ni	0.02 - 0.05 mils
Mild Steel	0.2 - 0.8 mils

Filter Section

Particulate filters of the type manufactured by Cambridge Filter Corporation, Flanders Filter Mills, Inc. and Mine Safety Appliances Company are used interchangeably and are mounted individually on the upstream side of individual 1000 CFM exhausters. No pre-filtering is provided.

In this installation, the air being filtered is a mixture of ventilation air and dissolver off-gas which has been passed through a caustic scrubber. Filter replacement is made on a scheduled basis based on their pressure drop build-up experience. As in other filter installations of this type, the penetration of particulate matter is less than 0.05% as measured by the standard DOP test. It was reported that filters were unaffected by 85 percent saturated air.

Oak Ridge National Laboratory - Oak Ridge, Tennessee - October 8, 1958

Oak Ridge was visited primarily to discuss the containment system of the ORR (Oak Ridge Research Reactor) and in particular the caustic packed column scrubber for removing iodine. However, we found that ORNL are or had been testing other iodine removal processes on a laboratory scale.

ORR Containment

J. A. Cox, ORNL	E. L. Etheridge, HAP0
T. E. Cole, ORNL	R. C. Walker, HAP0
F. T. Binford, ORNL	M. R. Schneller, AEC-HOO
A. L. Benson, AEC-ORO	

Under normal operating conditions, the ORR ventilation air is exhausted to the atmosphere via several small exhaust fans which are located in the external building wall. Upon indication of a release of fission products within the building, building sealing is provided by automatic closure of dampers in the ventilation openings as well as the two large truck entrance doors (can also be manually actuated from reactor control room). Containment is then accomplished by evacuating air from the building, through a filter-scrubber section, at such a rate that any air leakage through the building shell will be inward rather than outward.

Design of the building shell included caulking and taping all joints in the insulated metal siding, gaskets around all exterior access door jambs in addition to the automatic closures mentioned above. The filter-scrubber section

for the emergency exhaust air mentioned above consists of a fine filter of the CWS type (99.9 % efficiency) followed in series by a caustic scrubber. After passing through the filter-scrubber section, the air is discharged through a 250 foot stack.

Caustic Packed Column Scrubber⁽³⁾

E. Lamb, ORNL
A. L. Benson AEC-ORO

E. L. Etheridge, HAP0
M. R. Schneller, AEC-HOO

The ORR caustic packed column scrubber for removing an accidental release of fission product iodine from the reactor building ventilation air was designed for a decontamination factor* of 1000. The specifications of the scrubber are as follows:

Scrubber - Packed tower of ceramic berl saddles with air flow countercurrent to a descending stream of recirculated caustic solution.

Packing - 3/4 inches of 1 inch and 3/4 inches of 1-1/2 inch

Air flow - 5000 cfm

Caustic flow - 50 gpm, 5% NaOH solution

To obtain experimental data for the above scrubber, a model scrubber was designed using a scaled down factor of 0.0094. The specifications for the model scrubber were:

Scrubber - Same as above

Packing - 31 inches of 1/2 inch ceramic berl saddles, 0.0973 ft² cross sectional area

Air flow - 22.0 cfm

Caustic flow - 0.22 gpm, 5% NaOH solution

Trial runs of the model tower showed that it could not be operated at the scaled down air and caustic flow rates because of flooding conditions. By reducing the air and caustic flow rates it was possible to determine the efficiency of the test tower. The results of the test runs gave an average DF of 290. Extrapolation of the test data to the full size scrubber predicted a DF greater than 1000.

As a further check on the scrubber, efficiency tests were run on the full size scrubber. Iodine, traced with I131, was injected into the ORR exhaust duct approximately upstream of the scrubber. The average DF obtained from the series of tests was 100. In explaining the discrepancy between the two series of test data, it was felt that the air sampling technique may have been the cause of error.

Oak Ridge feels that a DF of 100 is about the maximum efficiency which can be obtained with a caustic packed column scrubber of reasonable height in removing dilute iodine concentrations from air at room temperatures. The pressure drop across the ORR scrubber is 3.8 inches of water at 6000 cfm air flow.

* Decontamination Factor = Concentration Entering

Activated Charcoal Bed(4)

W. E. Browning, ORNL
R. E. Adams, ORNL
A. L. Benson, AEC-ORO

E. L. Etheridge, HAP0
M. R. Schneller, AEC-HOO

To protect the ORR building and personnel from exposure to radiation from an accidental release of fission products from in-pile experiments on the reactor, all experimental equipment is to be designed so that the escaping fission products are released through a special off-gas system. The gas would be decontaminated and released to the stack. The off-gas system would be exhausted continuously, therefore the system would be handling large volumes of uncontaminated air for long periods of time but still remain in readiness for an accidental release.

An activated charcoal bed was tested to determine its efficiency. A full scale bed was designed for the special off gas system for an air flow of 1000 cfm. The specifications are as follows:

Bed size	2.75 feet, diameter
Depth	1.0 foot
Charcoal size	2-4 mesh
Air flow	1000 cfm
Air velocity	168 fpm
Pressure drop	2.8 inches of water

A scaled down model was then tested at an air velocity of 180 fpm with iodine, traced with I^{131} . It was determined that 99.95% of iodine was retained on the bed. The iodine appeared "fixed" on the surface, suggesting chemisorption rather than physical adsorption. No movement of the iodine down the column could be detected after 20 hours of air flow. Linde Molecular Sieve material was tested under similar conditions. The efficiency was approximately the same but a slow downstream transport of iodine was noted during the test.

Moisture did not appear to effect the adsorption of iodine on the charcoal surface. The maximum capacity of charcoal was not determined although 12 curies of iodine per gram charcoal was reported here at Hanford. (5)

One serious disadvantage of using a charcoal bed is the potential fire hazard. There is speculation that a possible surface concentration of radio iodine releasing decay energy plus the heat of adsorption may ignite spontaneously.

Fission Product Release from Molten Fuels Study

G. E. Parker, ORNL
A. L. Benson, AEC-ORO

E. L. Etheridge, HAP0
M. R. Schneller

Experiments in the controlled melting of irradiated fuel specimens, particularly alloyed fuel materials, are being conducted to determine the approximate percent of volatile fission products released during the meltdown. The irradiated sample is placed in an induction furnace in a controlled atmosphere. The atmosphere containing the volatile fission products is then swept into through a tube containing a filter and various traps for analysis.

Although meltdown experiments have been conducted for several years, the program is still in the preliminary stage. It is impossible to predict the percentage of particulate, volatile and noble gases released during a meltdown.

Of particular interest were the iodine results of two recently conducted MTR fuel element experiments in an air atmosphere. The analysis of the retained iodine is shown below:

<u>Retention Media</u>	<u>Run A</u>	<u>Run B</u>
5 μ paper (millipore)	91.7%	70.7%
0.8 μ paper (millipore)	0.6	2.1
NaOH Pellets	2.5	11.4
Charcoal (-195°C)	5.2	15.8

The analyses would indicate that at least 70% of the iodine was in the particulate form (IO_3^- and I^-) and could be retained on an absolute filter while the remaining 30% in the elemental form could be removed by adsorption, adsorption or chemical reaction. These figures are substantiated by the "quoted" figures of the Windscale incident in England where their filter system in the stack retained approximately 50 percent of the iodine released.

ORNL Graphite Reactor (X-10) Exhaust System

W. R. Casto, ORNL
R. V. McCord, ORNL

R. C. Walker, HAPC

The air coolant for the X-10 reactor is filtered in much the same manner as that proposed for Project CG-791; however, no provision for halogen treatment of the exhaust air is either presently included or anticipated. The inlet air to the reactor is filtered by American Air Filter Company PL-24 Airmat Filters loaded with 3/32" type G (Fiberglass) media. Air leaving the reactor is routed to the filter house which contains two filtering stages. The first or roughing stage consists of American Air Filter Deep Bed Filters, each loaded with one layer of Type FG-50 and one layer of Type FG-25 media. Following this is a bank of CWS type filters for fine filtering.

The exhaust air is discharged to a 200 foot stack by two 900 HP centrifugal compressors each of which deliver approximately 60,000 CFM at a static pressure of 51" w.g. Operating experience with the filter house has been very satisfactory. Roughing filters have been changed on the average of once every two years and fine filters have been replaced approximately each two and one-half years.

Homogeneous Reactor Experiment

S. Peterson, ORNL
A. L. Benson, AEC-ORO
R. A. McNess, ORNL

E. L. Etheridge, HAPC
R. C. Walker, HAPC
M. R. Schneller, AEC-HOO

ORNL experienced trouble in fission product iodine poisoning the copper oxide catalyst in the recombination system of the HRE. It was found that iodine in the off-gas system was depositing on the surface of the copper oxide and poisoning the catalyst necessary for the water recombination reaction of hydrogen and oxygen. To trap the iodine upstream of the recombination chamber, silver plated, stainless steel York mesh was installed in the form of a pad in the off-gas line. The York mesh was tested in the laboratory. At the HRE operating temperature and off-gas composition, the York mesh showed a 99 1/2 percent removal efficiency. At room temperatures, the silver plate mesh in HRE off-gas composition showed a low removal efficiency (50%).

Westinghouse Electric Corporation, Bettis Plant Atomic Division - Pittsburgh, Pennsylvania - October 9, 1958

W. T. Lindsay, WAPD
J. A. Wright, WAPD
W. E. McAllister, WAPD

E. L. Etheridge, HAPO
R. C. Walker, HAPO
M. R. Schneller, AEC-HOO

WAPD was visited to discuss the retention of iodine on absolute filters from a steam-air atmosphere. Some experimental work has previously been reported.⁽⁶⁾ This work was performed to demonstrate that when a leak in the coolant loop occurs and allows iodine to escape into the container the iodine can be retained by exhausting the dome air through a filter bank. Analyses of the loop coolant water have shown iodine present. It is thought that the iodine may be from the uranium impurity in the zirconium fuel element cladding. The experimental work indicated that approximately 90 percent of the iodine can be retained on absolute filters from a steam-air mixture. At the present there are no filters in the containment dome exhaust system.

Mine Safety Appliance Research Corporation is also performing some meltdown experiments for WAPD under a Bureau of Ships contract. The meltdown experiment is shown in schematic in Figure 2. The irradiated uranium specimen is heated to the melting point by induction heating. A steam-air stream is then passed over the molten uranium. The steam-air mixture is then allowed to escape into a large container (fall out chamber) and thence into the atmosphere. The analysis of the various gas streams showed that ~~25~~ ^{40-50 percent} percent of the iodine remained in the meltdown chamber, one to two percent was caught on the cold trap in the line to the atmosphere and the remainder settled out in the fall out chamber.

Another test being performed by MSA Research Corporation is the blowdown experiment. A schematic is shown in Figure 3. Radioactive iodine, ytterbium and sodium is added to the steam in the autoclave. The valve is opened to allow the steam to escape. The results of some of these tests have indicated approximately 95% ¹³¹I, 85% Na and 50% ytterbium settled out in the blowdown tank. The experiment has not been completed yet. A copy of the report will be forwarded to HAPO files upon completion of current tests.

Mine Safety Appliance Company - Pittsburgh, Pennsylvania - October 9, 1958

R. B. Evans
R. A. Bub
J. Summers

R. C. Walker, HAPO
E. L. Etheridge, HAPO
M. R. Schneller, AEC-HOO

The fine or "absolute" filters manufactured by MSA under the name of "Ultraire" are essentially similar to those manufactured by Cambridge Filter Corporation and Flanders Filter Mills, Inc. with one basic difference. In the MSA filters, no separators are provided between the folds of filtering media but rather, the folds of media are "pinched" together (somewhat like spot welding) with adhesive between the "pinches". It is claimed by the manufacturer that this modification to the basic filter design results in greater dust loading capacity, lower pressure drop characteristics and longer life.

Although there are no known independent tests to verify these claims, it would seem reasonable to expect some increase in performance characteristics of this nature since the space originally required for corrugated spacers is now available for additional air passage and/or dust loading. The rated characteristics of this filter are:

Efficiency - 99.97% minimum with standard 0.3 micron DOP test.

Initial ΔP - 0.9" (\neq 10%) w.g. @ rated capacity of 1000 CFM.

Life - Based on estimated dust concentration of 0.04 grains per 1000 cubic feet, element will have life of approximately 15,000 hours (625 days) and can be extended to as much as 45,000 hours (\sim 5 years) by adequate pre-filtering.

MSA also indicated they could provide a granular chemical filter which would remove iodine with a DF of 1000 or better. The pressure drop of the filter would be approximately 0.25 inches. The cost of the chemical filter would be \$150/1000 cfm. MSA will send the specifications of the halogen filters as soon as they are formulated.

Harvard School of Public Health, 55 Shattuck St., Boston 15, Mass. - October 10, 1958

Dr. L. Silverman, Harvard E. L. Etheridge, HAPO
C. Billings, Harvard R. C. Walker, HAPO
J. J. Fitzgerald, Harvard M. R. Schneller, AEC-HOO

Harvard was visited to discuss the water scrubber in removing radioactive iodine from an air stream because of the experience of Dr. Silverman in the field of air cleaning. However, we found that the Harvard University Air Cleaning Laboratory was conducting a series of experiments on removing radioactive iodine, principally I^{131} from waste gas streams for the Division of Reactor Development, AEC. Dr. Silverman is also a member of the ACRS.

In his capacity as an ACRS member, Dr. Silverman stressed the importance and the priority of installing the rear face fog spray system immediately. He also asked about the advisability of using demineralized water for the rear face fog spray system to eliminate the possibility of salt particle formation resulting from evaporation of the spray water. However, it was stated that a reactor incident of an energy magnitude which would evaporate a water flow of 300 gpm would probably destroy the integrity of the 105 Building also.

The Air Cleaning Laboratory have tested several media in removing stable iodine from an air stream at both elevated (300 C) and room temperatures (20 C). Some of the media are: Slag wool, silver plated slag wool, copper mesh beds, copper mesh beds with steam injected and silver plated copper mesh beds.

The slag wool proved to be ineffective removing iodine from air streams. The silver plated slag wool efficiency was approximately 98 to 99 percent at the start of the test but soon dropped below 90 percent. It was postulated that the drop in efficiency was due to difficulty in silver plating the inner surface of the dense slag wool and the high flow resistance through the bed. The air, after initial fixing of the iodine on the front surface of the bed, began channeling through the bed.

Copper ribbon (2 x 25 mils) beds were also tested and gave an efficiency of 99.9 % percent over extended test periods (25 hours). The resistance of a five-inch bed, with a packing density of 40 lbs/cubic foot, was approximately 1/2 inch water for a face velocity of 60 ft. per minute. Extensive testing must be made to determine the life of the collector beds.

Silver plated copper ribbon beds gave efficiencies of 99.9 % over extended test periods. The filter has a fail safe feature in that any stripping of the silver plating would leave the copper filter surface available as the collector. The beds have been exposed to hydrogen sulfide and nitric acid atmospheres and retested with no apparent reduction in efficiency.

It appears that silver plated copper ribbon beds is the best media for removing iodine from air streams tested at Harvard to date. Tests have been run with face velocities from 60 to 240 fpm with no loss in efficiency. Pressure drop is very low. A two inch bed having a packing density of 27 lbs/cubic feet had a pressure drop of only 0.1 inch of water at a face velocity of 60 fpm.

It should be remembered that these tests were run with stable iodine and at higher concentrations, 80-40 ppm) that are expected from our most probable reactor incidents (0.5 - 1.0 ppm). The Harvard Lab plans to corroborate these data using I¹³¹.

Brookhaven National Laboratory - Upton, L.I., New York, October 13, 1958

B. Manowitz, BNL
M. Steinberg, BNL
T. Sheeham, BNL
R. W. Powell, BNL

E. L. Etheridge, HAP0
R. C. Walker, HAP0
M. R. Schneller, AEC-H00

BNL did not have any experimental work on the problem of iodine removal systems. They did have some absorption data on xenon and krypton. They have found increasing solubility of the noble gases in kerosene and other non-polar organic liquids. These solubility capacities in kerosene were 2-3 cc/cc for xenon and 1/2 cc/cc for krypton.

BNL's air cooled reactor is quite similar to the ORNL X-10 reactor; however, the philosophy and method of handling ventilation air is slightly different. Inlet air to the reactor is filtered by American Air Filter Deep Bed Filters with FG-100 media (essentially same as roughing filter in ORNL filter house) and air exhausted from the reactor is filtered by a specially designed woven glass cloth filter (Dollinger). This latter filter is rated at 95 percent efficiency for five micron particles. While this is somewhat below our proposed specification (and ORNL's existing standards) they have reportedly maintained the release of particulate to the environs within tolerable limits. They are, however, planning to replace these filters with ones of the standard CWS type, not so much from the standpoint of improved filtering performance, but because they feel that the CWS filter has some additional degree of halogen collection. No system is included in either the BNL or ORNL reactors which is specifically designed for halogen removal.

Finally, the exhaust air is discharged to the stack by a series of centrifugal compressors operating in parallel, each rated at 35,000 CFM at 92 in. w.g. Fan drives are 1500 HP motors operating at 3600 RPM. Because their existing system characteristic is more on the order of 50 in. w.g., plans are being made to modify the fans accordingly. In particular, they are anxious to reduce the motor speed due to excessive maintenance experience.

BNL's experience with ruptured natural uranium fuel elements have indicated that most of the fission products released have been found in specs (micron size). The fission products appear to be gathered on an uranium oxide particle.

Schuyler Manufacturing Corporation - North Arlington, New Jersey - October 14, 1958

E. E. Reynolds
C. C. Cone

E. L. Etheridge, HAPO
R. C. Walker, HAPO

The Schuyler entrainment separator is a compacted bed of a wire net mesh. The water droplets in the air are removed by impinging on the surface area of the mesh. The tiny droplets collect to form a large droplet which then falls into the collection sump. The specifications of the entrainment separator are:

Material	304 stainless steel
Density	9 lbs/ft ³
Surface area	85 ft ² /ft ³
Wire diameter	0.011 inches
Entrainment Loading	1600 lbs water/(hr)(ft ²)
Bed thickness	6 inches
Face velocity	3-17 fps
Pressure drop	0.5 - 2 inches
Minimum Efficiency	99.9

It was recommended that the separator be placed in the horizontal position as variations greater than 10 degrees from the horizontal position causes flooding in the lower section of the separator and the possibility of re-entrainment through the bed with a consequent reduction in efficiency). (The ANL horizontal caustic scrubber entrainment bed is mounted on a 45 degree plane but with baffles preceding the bed. There was no visible indication of water droplet on the downstream side.) The cost of a six inch entrainment separator bed with supporting grid work is \$14.25/sq. ft, face area.

Schuyler was also asked about silver plated copper ribbon beds. They fabricate a similar round wire mesh for shock absorber joints for the Air Force and for shields in electronic tubes. It would be a simple matter to substitute flat ribbon in the fabrication. The cost of a 24" x 24" x 2" silver plated copper flat wire, (0.002 x 0.018 inch) bed, having a packing density of 27 lbs/cu. ft. was quoted at \$44.50. The capacity of the pad is 1000 cfm.

Cambridge Filter Corporation - Syracuse, N.Y.

P. M. Engle, Cambridge Filter Corp. R. C. Walker, HAPO
D. H. Northrup, Cambridge Filter Corp.

The Cambridge "Absolute" Filter is the one currently being utilized at HAPO on the basis of extensive moisture and burning tests which have been conducted by CPD. Although these tests are considered to be more severe than would be required for the CG-791 application, Mr. Engle feels that this particular filter (Cambridge Type "G") would most nearly meet our requirements for our fine fitter.

Of particular interest is a roughing filter manufactured by Cambridge under the name "Aerosolve". The initial cost of these filters is somewhat higher than the American Air Filter PL-24; however, the "Aerosolve" has several features which are very desirable.

- a. The Aerosolve filter consists of a permanent metal frame and one of three cartridge, all of which are interchangeable and easily replaceable. These cartridges are presently designed to provide efficiencies of approximately 95%, 85% or 35% but could be manufactured to any intermediate value. Thus it is possible to change to a more or less efficient cartridge as operating experience dictates.
- b. Since the entire cartridge is replaced when filter become loaded, there is less change of spreading contaminated material. In most other types of roughing filters, the filter unit consists of a metal frame and a removable cartridge; however, the cartridge framework must be stripped of the dirty filter and reloaded manually. This would obviously be an inconvenient method for replacing contaminated filters.
- c. The dust loading of the "Aerosolve" is much higher than the PL-24 which would imply less frequent filter changing.

Data on various Cambridge Filters are given below:

	Cambridge (Absolute)	Aerosolve 35	Aerosolve 85	Aerosolve 95
Efficiency (%)	99.95 /	30-40	80-85	9-95
Initial pressure				
Drop (in. w.g.)	1.0	0.16	.022	0.35
Recommended final				
Pressure drop				
(in. w.g.)	2.0	0.50-0.60	0.60-0.65	0.70-0.80

Manufacturer estimates that life of absolute filter will be extended about 1.5 times by using Aerosolve 35 pre-filter and about 3.0 times by using Aerosolve 85 pre-filter.

E. L. Etheridge

RC Walker

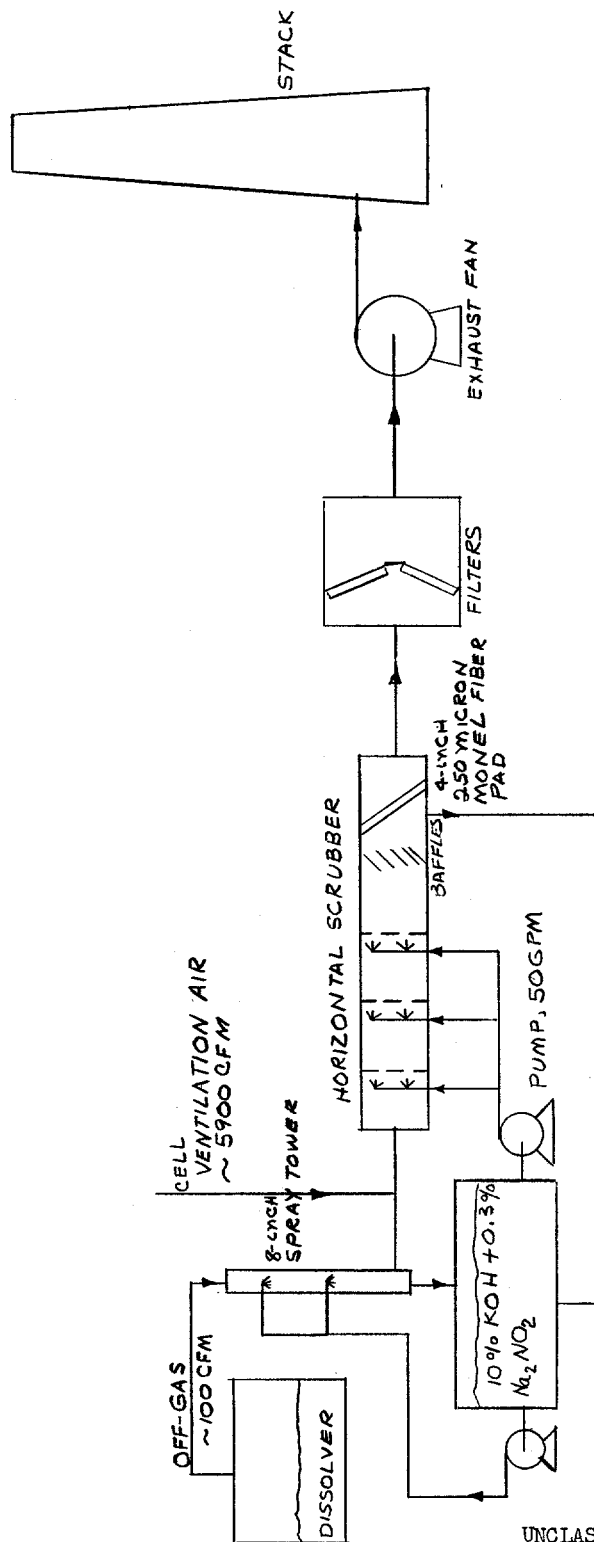
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EL Etheridge:RC Walker:bsm

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5. HW-26113, "Removal of Iodine Vapor from Gas Streams by Sorption on Charcoal", J. W. Finnigan, et. al., November 4, 1952.
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FIGURE 1. ANL OFF-GAS EXHAUST SYSTEM



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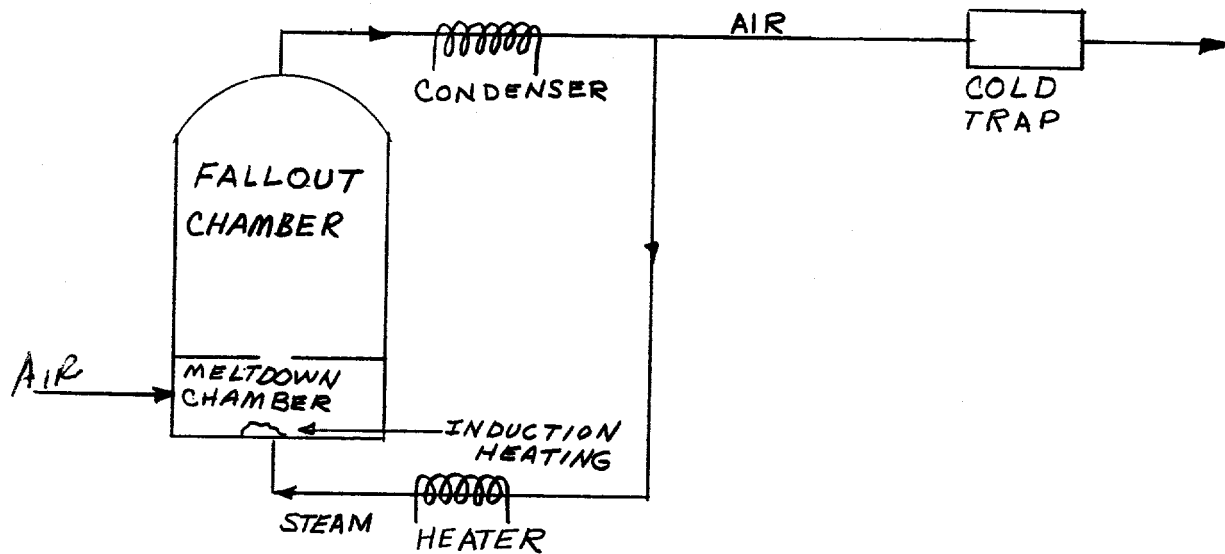


FIGURE 2. MSA MELTDOWN EXPERIMENT

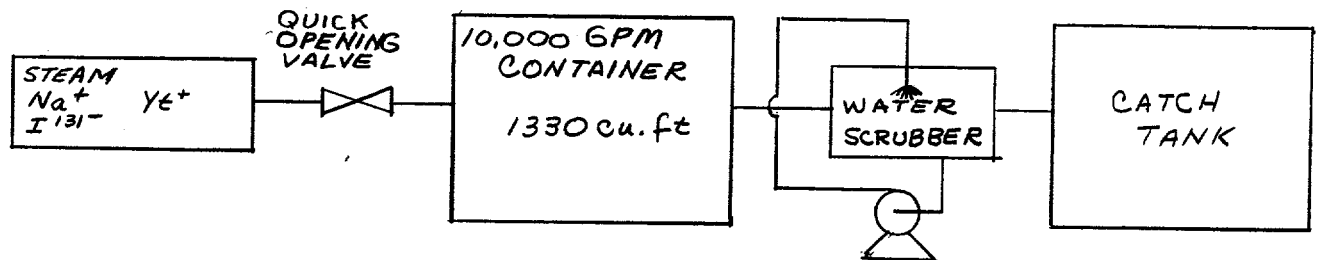


FIGURE 3. MSA BLOWDOWN EXPERIMENT